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Study of electrical conduction mechanism of succinic acid doped glycine pellet

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Abstract The electrical conductivity of succinic acid doped glycine pellet has been measured by studying the I-V characteristics at various temperatures in the range 313K-353K. The results are presented in the form of I-V characteristics and analysis has been made by interpretation of Poole- Frenkel, Fowler- Nordheim, Schottky, log(J) *versus* T plots, Richardson and Arrehenius plots. It is observed that the conduction mechanism in the present case is a cooperative process, with Poole-Frankel type in low field range and Fowler-Nordheim mechanism in high field range

Keywords Succinic acid glycine pellet, conductivity

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1. Introduction

The bioelectret state plays an important role in many biological phenomena and is a universal property of biomolecule. The bioelectret state manifests in a number of biological materials. Biomolecules are related to each other and interact in a kind of molecular game of logic. The typical physical properties of the biomolecules enable them not only to serves as building blocks of the intricate structure of cells, but also to participate in their dynamics, self-sustaining transformation of energy and matter [1].

In the present years, a great deal of interest has been centered on the study of physical properties [1, 2] of amino acids because it may be helpful in understanding both physical and chemical properties of peptides, proteins and enzymes, as amino acids are the basic building blocks of these biological macromolecules [1]. Glycine (aminoacetic acid), NH_2-CH_2-COOH is a natural aminoacid which serves as a precursor in the biosynthesis of purines, porphyrins and creatine and is a component of the active site in many enzymes [3]. Anagnostopoulou and Pissis [4] and Brideli *et al* [5] have worked on biopolymers like poly peptides and proteins. Mishra and Nath [6] measured the dc-electrical conductivity of glycine to study the mechanism of electrical conduction and results are analysed in terms of Schottky – Richardson mechanism which is primarily responsible for the conduction.

Belsare and Deogaonkar [7] have reported the electrical conductivity of iodine-doped polyblend films of polystyrene (PS) and polymethyl methacrylate (PMMA). The electrical conductions in iodine-doped polystyrene (PS) and polymethyl metharcrylate (PMMA) have already been reported [8,9]. Japanese scientists [10] have been particularly active in early research and development of these devices with work on natural and synthetic polymers. Most of the early polymer electret work in the U.S., have been focused on using the polyelectric response for electromagnetic radiation detection [11, 12].

In the case of organic solids where the conductivity due to electrons excited from valence band to conduction band [13, 14] is negligible, a complex conduction behaviour [14, 15] has been explained usually in terms of electron emission from cathode *i.e.* Schottky-Richardson mechanism [16] or by electron liberation from the traps in the bulk of the material *i.e.* Poole-Frenkel mechanism [17]. Possibility of tunneling [18], spacecharge limited conduction [19] *etc.*, have also been discussed in the literature.

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In the present paper, dc conductivity of succinic acid-doped glycine was measured to identify the mechanism of electrical conduction. It is shown how the I-V data of the sample can be used to arrive at possible conclusions.

2. Experimental details

2.1 Preparation of samples :

Glycine and succinic acid used in the present investigation, was the standard grade product supplied by HI-MEDIA Laboratories Pvt. Ltd., Mumbai, in the powder form. The homogeneous mixture of 94% glycine (0.1034g) and 6% succinic acid (0.006g) was prepared by using a shell and mortar and successively rubbing together for hours so that an intimate mixture is obtained. This intimate mixture was poured in a pellet die which was then subjected to a uniform pressure of 3 tons/cm^2 using a KBR Press. The pressure was applied for three minutes. A fine uniform shining pellet of the sample mixture was obtained with perfectly smooth parallel faces. Both the surfaces of the pellet were coated with quick drying silver paint supplied by Eltecks Pvt. Ltd., Banglore to ensure good electrical contacts. The coated sample pellet was subjected to uniform heating at a temperature of 100°C in the furnace.

The X-ray powder diffractogram of pure glycine and glycine doped with 6% succinic acid, have been recorded from a Phillips automated diffractometer (RSIC, Nagpur). Figure 1 shows both the diffractograms. As the strongest lines (100% peaks) of α-glycine (d = 2.990Å [$h\,k\,l$] = [0 4 0]) and β -glycine (d = 4.91 Å [$h\,k\,l$] = [0 0 1]) are not found in the diffractogram, it is inferred that α and β-glycine are absent in the sample. However, almost all peaks (except weak lines) index as those of γ -glycine or γ - succinic acid. Thus the sample is inferred as γ -glycine doped with 6% succinic acid. Table 1 shows the indexing of the diffractogram.

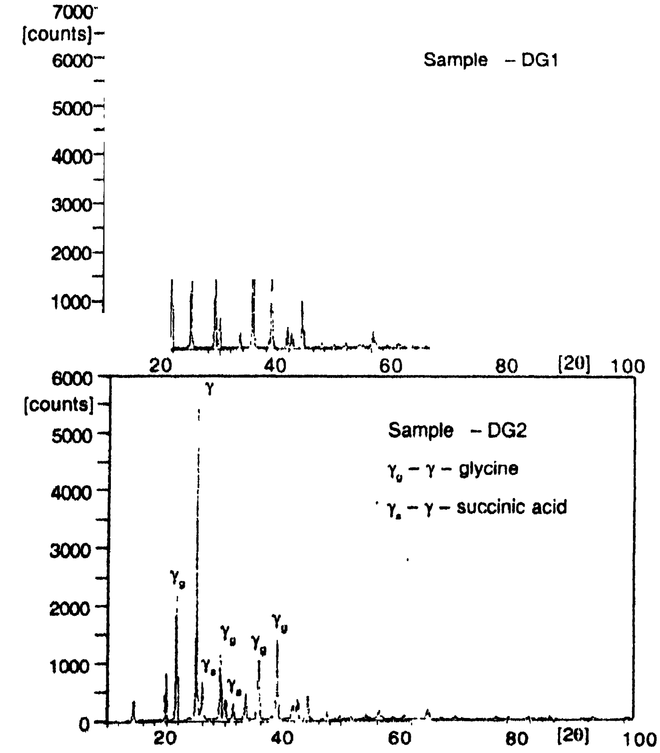

Figure 1. X-ray diffractogram of sample DG1 and DG2.

Table 1. Indexes of the diffractogram.

d (Å) values					
Standard	Observed	h	k	l	
4.420	4 418	0	2	0	γ_s
4 070	4.086	1	0	1	γ_g
3 51	3 528	1	1	0	γ_g
3 397	3 416	1	1	0	γ_s
3 054	3.054	2	0	0	γ_s
2.832	2 844	1	2	0	γ_s
2 500	2 504	1	0	2	γ_g
2.300	2.308	2	1	0	γ_g
2.166	2 167	1	1	2	γ_g
2 030	2 035	3	0	0	γ_g
1.900	1.908	3	0	1	γ_g
1.630	1 633	3	0	2	γ_g
1 610	1.619	3	1	1	γ_g
1 520	1 526	4	0	0	γ_g

γ_g = γ - glycine
γ_s = γ - succinic acid

2.2 Measurements :

Thermostatically controlled furnace supplied by Tempo Industrial Corporation, Mumbai was used for heating purpose. A mercury thermometer with an accuracy of ± 1°C was used for recording the temperatures. The regulated power supply supplied by Nupur, India was used as the voltage source, while the current was recorded by using highly sensitive Pico-Ammeter (Model DPA 111) with accuracy ± 0.2% supplied by Scientific Equipments, Roorkee.

The sample pellet coated with silver electrodes was sandwiched between two brass–electrodes (diameter 2.4cm) of the sample holder specially fabricated in the laboratory. This formed the Metal- Insulator–Metal (M–I–M) system, which was placed in a furnace. The current (I)- voltage (V) measurements were done at various temperatures from 313K to 353K.

3. Results and discussion

The log I–log V plots of succinic acid doped glycine pellet at temperatures 313K, 323K, 333K, 343K and 353K are shown in Figure 2. The current increases non linearly with the applied voltage and does not follow power law $I = k\,V^m$, where k and m are constants. The current in the beginning, at low values of voltages, increases at a faster rate while it is being slowed down at higher values of voltages. Figure 1 indicates that (i) the current at a constant temperature increases with applied voltage, (ii) the current at constant applied voltage increases with temperature.

The mechanism operative in present case is discussed in the light of the Poole -Frenkel, Fowler–Nordheim, Schottky, ln (J) *versus* T plots, Richardson and Arrehenius plots.

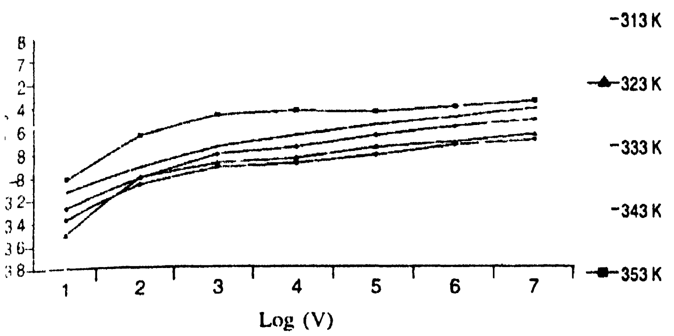

Figure 2. Current volatage characteristics

3.1 Poole-Frenkel mechanism :

The current–voltage relationship for Poole-Frenkel mechanism is expressed as

$$J = B\exp\left((-\varphi/kT) + \beta_{PF} E^{1/2}\right),$$

$$\beta_{PF} = (e/kT)(e/\pi\varepsilon\varepsilon_0 d)^{1/2} = \text{Constant} \tag{1}$$

and predicts a field dependent conductivity as

$$\sigma = \sigma_0 \exp\left(\beta_{PF} E^{1/2}/2kT\right) \tag{2}$$

or $$\ln\sigma = \ln\sigma_0 + (\beta_{PF}/2kT)E^{1/2}. \tag{3}$$

So that $\ln\sigma$ *vs* $\sqrt{E}$ plots *i.e.* the Poole-Frenkel plots as predicated by eq. (3) are linear with the + *ve* slope.

In the present case of glycine doped with succinic acid, the $\ln\sigma$ *vs* $\sqrt{E}$ graphs shown in Figure 3 :

i) are linear with + *ve* slope for low values of $\sqrt{E}$, at higher temperatures. This indicates a thermal excitation of trapped electrons in the conduction band. This is further assisted by lowering of trap depths by the applied field.

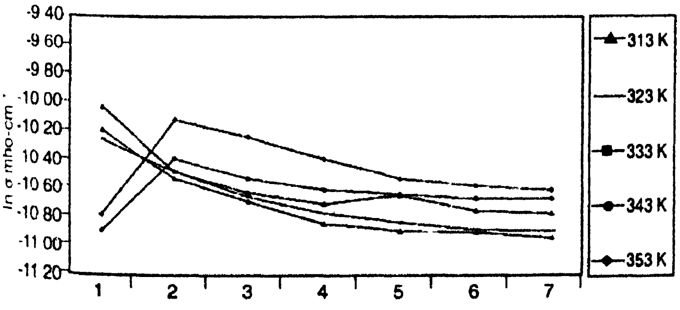

Figure 3. Poole Frenkel plots $\sqrt{E}$ (volt – cm^{-1})$^{1/2}$.

ii) in the lower range of E, but at lower temperature, the graphs are straight lines with a–ve slope indicating absence of PE mechanism in this range.

iii) beyond a particular value of $E(\sqrt{E} = 2V/m)$, the curves are straight lines with again –*ve* slope indicating absence of PE mechanism in this region, for higher as well as lower the temperature.

3.2 Fowler–Nordheim mechanism :

The Fowler – Nordheim relation (18) for current density is

$$J = AV^2 \exp(-\varphi/v) \tag{4}$$

So that, $\ln\frac{J}{V^2} = \ln A(-\varphi/v)$

And the $\ln J/V^2$ *versus* $1/V$ plot is expected to be of linear relation with a –*ve* slope.

(i) In the present case, the $\ln(J/V^2)$ *versus* $1/V$ plot for the sample are presented in Figure (4) which are nearly straight lines with –*ve* slope for higher values of (and of course E) indicative of the process of tunneling current as suggested by $F-N$ relation.

(ii) The curve are almost horizontal (with a –*ve* slope) for lower values of V indicating absence of $F-N$ mechanism.

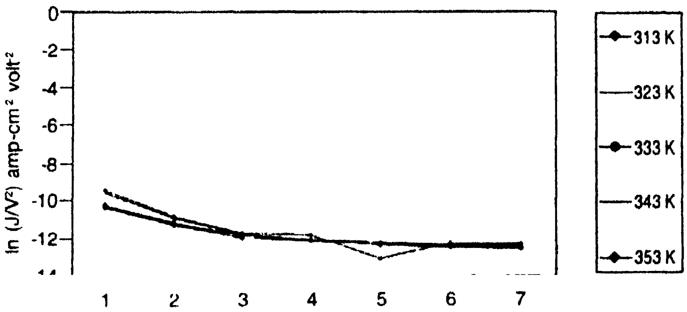

Figure 4. Fowler Nordheim plots 1000/V (volts)$^{-1}$

3.3 Schottky plots :

The Schottky – Richardson current voltage relationship is expressed as

$$J = AT^2 \exp\left(\frac{-\varphi_s}{kT} + \beta_{SR} E^{1/2}\right) \tag{5}$$

$$\beta_{SR} = (e/kT)(e/4\pi\varepsilon\varepsilon_0 d)^{1/2}$$

and hence $\ln J = \ln AT^2 - \varphi_s/kT + \beta_{SR} E^{1/2}$ and that $\ln J$ *versus* $\sqrt{E}$ plot should be a straight line with a +*ve* slope. In our case, the $\ln J$ *versus* $\sqrt{E}$ plots are nearly horizontal lines as shown in Figure 5 specially at higher values of E, indicating absence of Schottky mechanism. This shows that thermal activation of electrons over the metal insulator interface does not occur even with the added effects of the applied field which should have reduced the height of the barrier. *i.e.* Schottky mechanism is not applicable to the present case.

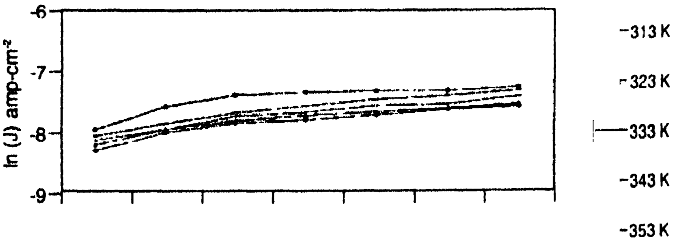

Figure 5. Schottky plots /E (volt cm^{-1})$^{1/2}$

3.4 Richardson mechanism :

The ln J/T^2 *versus* $1/kT$ plots (Figure 6) are referred to as the Richardson plots and the mechanism expects a linear relation with *-ve* slope. Here again, the slope is extremely small in almost all cases showing no applicability of this mechanism for conduction.

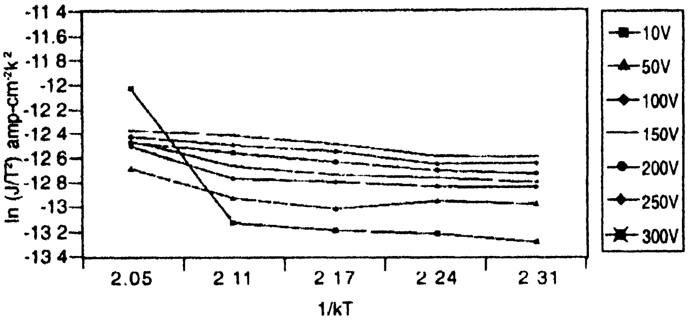

Figure 6. Richardson plots.

3.5 Arrehenius plots :

The ln σ *vs* 1/T Plots (Figure 7) at all values of applied voltage show parallel straight lines with a *-ve* slope for all values of applied field. This enables us to calculate the activation energy for conduction which has been found out to be 0.21 eV. This is in good agreement with the reported order of magnitudes.

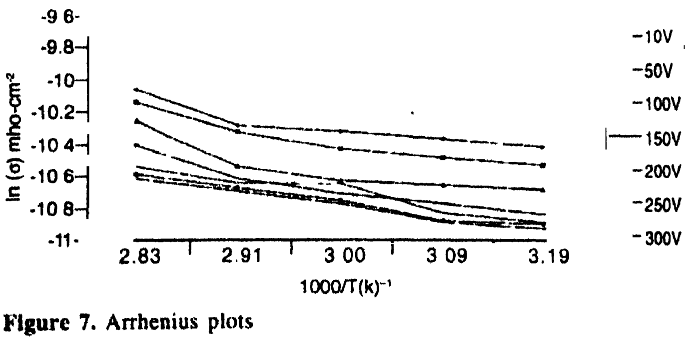

Figure 7. Arrhenius plots

3.6 Current density vs temperature plots :

The temperature dependence of current is presented in the form of ln *J vs T* plots (Figure 8) and that the increment in current due to rise of temperature is rather poor. The strong temperature dependence is suggestive of Schottky – Richardson mechanism, which is not observed in this case. With rise in temperature, succinic acid may start dissociating, first forming monocarboxylate ion (at lower temp) and increase in conductivity may be attributed to this phenomenon.

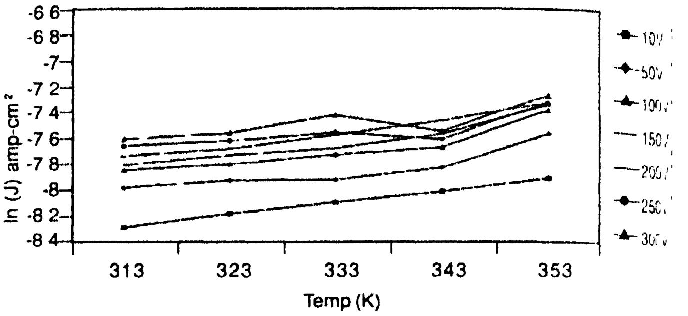

Figure 8. Current density *vs* temperature plots.

4. Conclusion

It appears that the mechanism of electrical conduction is a cooperative process here, with

i) Poole-Frenkel mechanism indicating thermal excitation of trapped electrons, into the conduction band *via* field assisted lowering of trap depth by the applied field at higher temperatures.

ii) Fowler – Nordheim mechanism which is indicative of the process of tunneling current at higher values of V. The study of conduction mechanism in pure glycine by Mishra and Nath [20] shows Schottky Richardson mechanism responsible for conduction.

In the present case of glycine doped with succinic acid there is a change to Poole – Frenkel mechanism in the lower range of field E, which subsequently changes to Fowler - Nordheim mechanism for higher range of E. The succinic acid provides trapping centers, which are subsequently thermally excited to conduction band *via* field assisted lowering of trap depths, in the lower range. In the higher range, conduction may be due to tunneling of electrons across the barrier causing Fowler Nordheim mechanism. Thus, dopants like succinic acid do play a role in altering the conduction mechanism in case of biopolymer like glycine.

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